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The structure of Sb-terminated GaAs(001) surfaces

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Abstract

We have studied the structure of Sb-terminated GaAs(001) surfaces using reflection high energy electron diffraction, X-ray photoelectron spectroscopy, and scanning tunneling microscopy (STM). Clean, As-terminated (2×4) surfaces were prepared by molecular beam epitaxy and then exposed to Sb₄ at 490°C, producing a (2×8)-reconstructed surface terminated with ~ 1 ML of Sb. Re-heating such a surface to 460°C in vacuum returns the surface to a (2×4) reconstruction with approximately 0.5 ML Sb remaining. STM reveals a complex, but well-ordered structure on the (2×8) surface for which a tentative model is proposed. On the (2×4) surface, our results clearly show that each unit cell is terminated by *three* dimers, with two-thirds Sb dimers and one-third As dimers. These results contrast with previous proposals that the Sb-induced (2×4) surface is terminated solely by one or two Sb dimers. © 1999 Elsevier Science B.V. All rights reserved.

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The structure of Sb-terminated GaAs(001) surfaces is of interest for a variety of reasons. A number of novel III-Sb semiconductor-based electronic devices are under development [1], and many of these devices are fabricated on semi-insulating GaAs(001) substrates by molecular beam epitaxy (MBE). As such, Sb-terminated GaAs represents the initial surface on which subsequent heteroepitaxial layers are grown. Sb-terminated GaAs is also the starting surface for the growth of self-assembled III-Sb quantum

dots on GaAs for potential use in optoelectronic devices [2–5]. In addition to forming an important interface in devices, Sb is also being considered for use on GaAs as a passivating surface film [6], and as a surfactant for heteroepitaxial growth [7,8].

Previous studies of Sb-terminated GaAs(001) have started with clean, As-terminated (2×4)-reconstructed surfaces that had been covered with multiple layers of Sb at low temperature and then annealed in vacuum to gradually desorb the Sb film [6,9–13]. With increasing temperature, reflection high energy electron diffraction (RHEED) patterns evolving from (1×4) to (1×3) to (2×4) have been reported for the resulting Sb-covered surfaces [6,9,14]. Low-resolution scanning tunneling microscopy (STM) images of surfaces with

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(1×4) and (1×3) periodicity showed little long-range order, providing little basis for structure determination [11]. Much more effort has been expended to determine the structure of the Sb-(2×4) surface, including analysis via high-resolution core-level X-ray photoelectron spectroscopy (XPS) [9], STM [11], reflectance anisotropy spectroscopy (RAS) [12], X-ray standing wave (XSW) measurements [10,13], and theoretical calculations [12,15–17]. To our knowledge, all the structures proposed for the (2×4) to date include *only* Sb surface dimers (no As surface dimers). Although XPS results have led to a proposal of three Sb dimers per (2×4) unit cell [9], and the STM study proposed a single dimer [11], the most recent experimental study concluded that the surface is terminated with two Sb dimers in a structure analogous to the As-terminated GaAs(001)- $\beta 2(2 \times 4)$ reconstruction [13]. This conclusion is at odds with the most recent calculations, which found the most likely structure to be one with a single Sb dimer on the surface [16].

In this Letter we describe our study of the structure of Sb-terminated GaAs(001) surfaces prepared by MBE and characterized with RHEED, STM, XPS, and X-ray diffraction (XRD). Rather than depositing thick films at low temperature, we first examined the reaction of Sb₄ at normal growth temperatures, where we observe a (2×8) structure. Annealing this structure in vacuum creates a (2×4) surface analogous to those studied previously. Our STM results clearly show that the Sb-terminated (2×4) surface has a structure different from that previously proposed.

The experiments were performed in a multi-chamber ultra-high vacuum (UHV) facility consisting of multiple interconnected MBE and surface analysis systems [18]. Surface structure was monitored during growth and Sb₄ exposure via RHEED. Prior to Sb exposure, 1 μm -thick GaAs buffer layers were grown under As-rich (2×4) conditions at 580°C with 30 s interrupts every 90 s. The surface was then cooled to $\sim 490^\circ\text{C}$ while simultaneously reducing the As₂ flux, producing a well-ordered (2×4) RHEED pattern stable under vacuum. To produce an optimal Sb-terminated (2×8) surface, the Sb₄ flux was slowly brought

up to about 1 ML/s over a period of 10 min, at which time the sample temperature was reduced to about 430°C. When the (2×8) RHEED pattern looked optimal, the Sb flux was terminated and the sample was allowed to cool to room temperature. To create the Sb-covered (2×4) reconstruction, an Sb-(2×8) surface was slowly reheated to 460°C in UHV over the course of about 10 min. After cooling, samples were transferred in UHV to a surface analysis chamber for characterization by either STM or XPS [19].

In order to obtain an absolute measure of the amount of Sb on these surface reconstructions, we performed XRD on a specially grown GaAs–Sb superlattice (SL) [20]. A 40 period SL was grown by MBE, with each period composed of 9 ML (26 Å) of GaAs grown under (2×4) conditions and then terminated with an Sb-(2×8) surface. Diffraction profiles (θ – 2θ scans) for this structure include superlattice satellite peaks which can be used to determine the superlattice period. Using Bragg's law and the position of the zeroth-order satellite peak, we calculated the average GaAs_{1–x}Sb_x lattice constant in the structure, which in turn was used to obtain the Sb concentration in the structure. Assuming all of the Sb on the (2×8) surface is completely incorporated into the SL, which we believe to be the case, this analysis leads to an Sb coverage of between 0.8 and 1 ML (depending on growth temperature) [20]. If all of the surface Sb does not remain, this is a *lower* bound on the coverage. From the integrated intensity of the XPS Sb 4*d* core level, we find the (2×4) surface has approximately half as much Sb as the (2×8), or approximately 0.5 ML. Note that this is the same absolute coverage for the Sb-(2×4) surface recently reported based on Rutherford backscattering [13].

In order to consider the structure of the Sb-terminated GaAs(001) surfaces, it is necessary to be clear about the structure of the initial As-terminated (2×4) reconstruction As illustrated in Fig. 1, our growth procedures produce large, well-ordered terraces with few islands or As vacancies. Our high-resolution STM images clearly show atomic-scale structure consistent *only* with the $\beta 2(2 \times 4)$ model for this reconstruction — rows of As dimer pairs (0.5 ML As) on top of 0.75 ML of

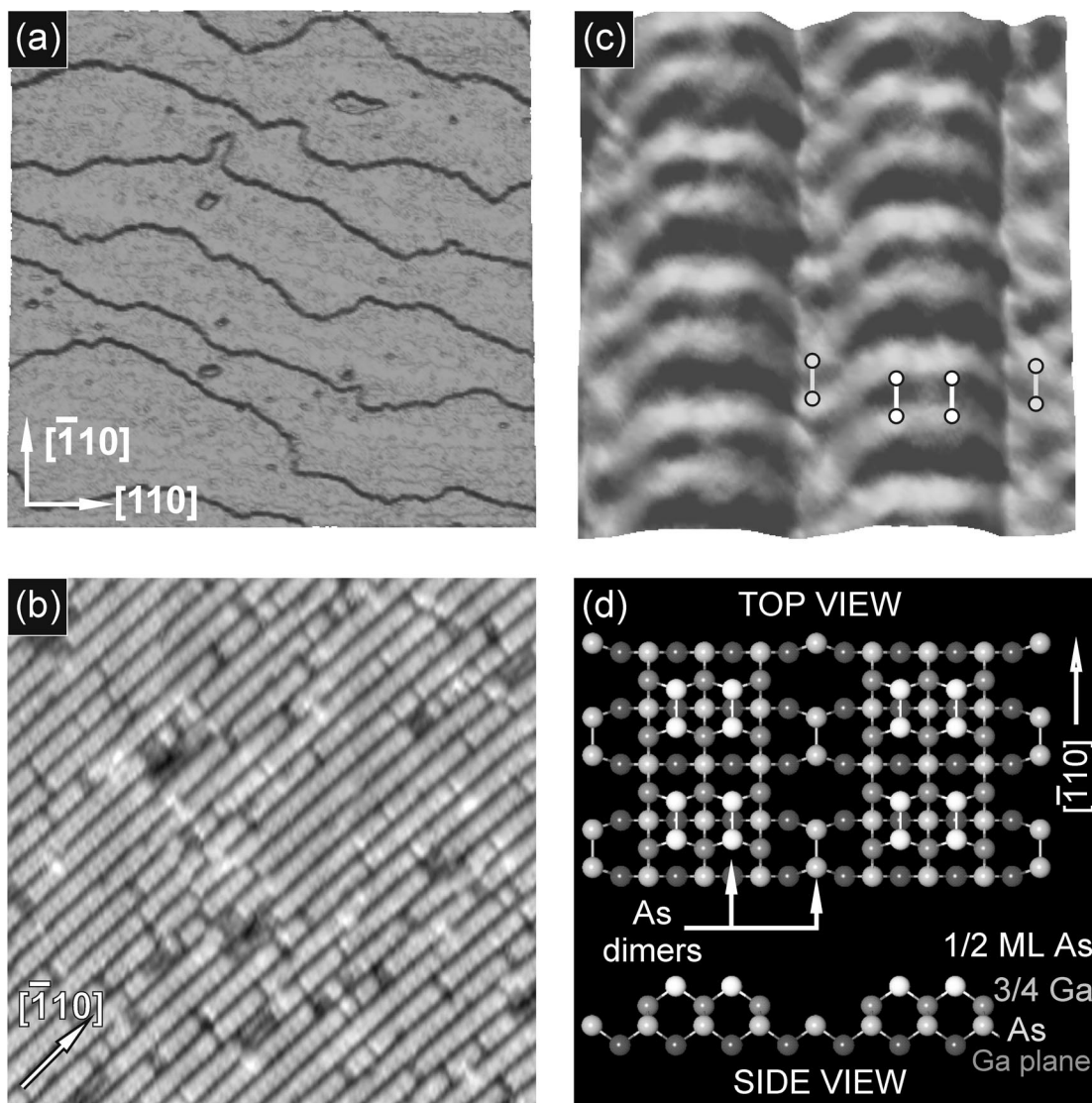


Fig. 1. (a) Large-area STM image (3D-rendered) showing the topography of an As-terminated GaAs(001)-(2 × 4) surface prepared by MBE with frequent growth interrupts (1.6 μm × 1.6 μm). (b) Higher magnification gray-scale image, 37 nm × 37 nm, showing the dimer rows of the reconstruction (filled states, 3.5 V, 30 pA). (c) 3.3 nm × 3.3 nm rendered view of the atomic-scale topography (filled states, 3.0 V, 0.1 nA). Note that the features associated with both the rows of As dimer pairs and the single row of As dimers in the layer below are just resolved. (d) A ball-and-stick model of the β₂(2 × 4) reconstruction.

Ga and separated by a row of As dimers in the layer below — confirming a variety of recent experimental and theoretical results [21]. To our knowledge, this is the first direct observation of the structure both on top of and between the As dimer rows, which is required to distinguish among the many models proposed for this reconstruction.

STM images of the Sb-induced GaAs(001)-(2 × 8) surface are shown at various magnifications in Fig. 2. The reconstruction has a grating-like appearance at lower magnification (Fig. 2a), with long, straight rows running along the $[\bar{1}10]$ direction that have a 32 Å period (× 8) along $[110]$. Although the rows are occasionally interrupted by

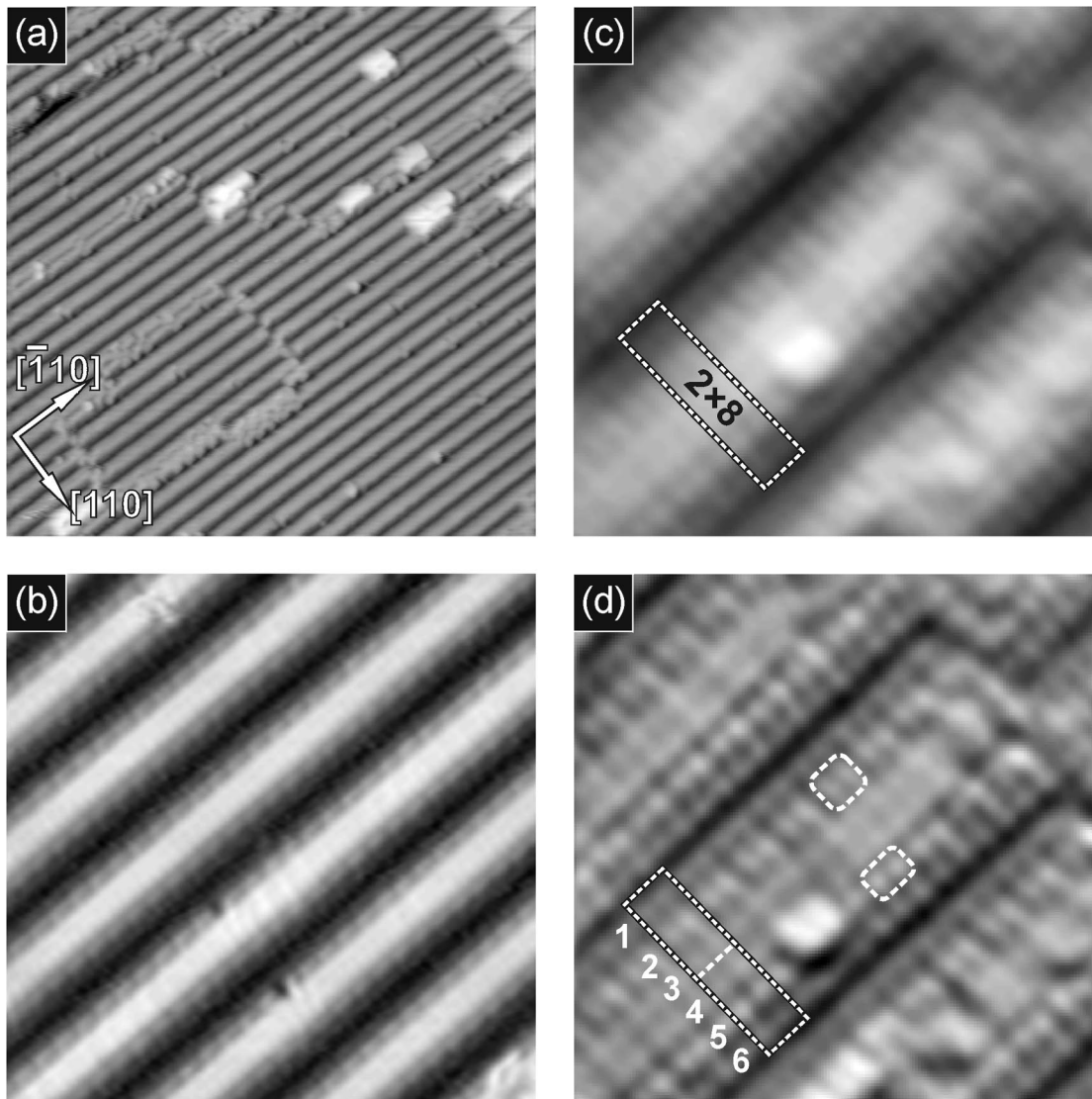


Fig. 2. Filled-state, gray-scale images of the (2×8) -reconstructed surface created by exposing a $\text{GaAs}(001)\text{-}\beta 2(2 \times 4)$ surface to Sb_4 at 490°C . The image sizes and tunneling conditions were as follows: (a) 82 nm, 3.3 V, 0.2 nA; (b) 17 nm, 3.3 V, 0.2 nA; (c) 8 nm, 2.8 V, 2 nA. (d) The image shown in (c) after high-passed filtering to accentuate the local height variations. The dashed line within of the unit cell indicates the midpoint. The different widths of rows '2' versus '5' are highlighted.

small islands or domain boundaries (shifts along $[\bar{1}10]$), on the most well-ordered surfaces individual rows frequently run uninterrupted across the length of the terrace (100–500 nm). At higher magnification, each row has a three-tiered appearance, as displayed in Fig. 2b and c. Measured from the bottom of the groove between the rows, the height of the first tier on either side is characteristi-

cally 1.5 \AA , the second tier 2.5 \AA , and the top tier 3.5 \AA . Each tier has a $2 \times$ periodicity along $[\bar{1}10]$ associated with dimer-like structures, accounting for the (2×8) periodicity observed in diffraction.

Because the atomic-scale features are difficult to discern with a gray scale that is linear in height, we have applied a high-pass filter to the data to convert the gray scale to a representation of the

'local' height. As seen in such a filtered image in Fig. 2d, it appears there are six atomic-scale rows between the grooves in each (2×8) unit cell. The two rows on top of the structure in the middle of the unit cell have a diffuse look, with a very weak $2 \times$ periodicity along $[\bar{1}10]$. In contrast, the other rows within the structure are clearly dimer-like, with their dimer bonds oriented along $[\bar{1}10]$. The appearance of *six* rows per $\times 8$ is surprising, given that a structure commensurate with the GaAs substrate would nominally be expected to have *seven* rows between the ~ 4 Å-wide ($\times 1$) grooves. Although at first glance this structure looks symmetrical with respect to the center of the unit cell, close inspection reveals a subtle but *reproducible asymmetry*. As seen in Fig. 2d, the node between the two middle rows occurs about 0.5 Å from the center of the unit cell (to the lower-right of the dashed dividing line). Similarly, the row to the upper-left of the top (labeled '2') is characteristically wider than the corresponding row on the other side (labeled '5'). This subtle asymmetry is more apparent under some tunneling conditions than others, but has been observed in both directions with respect to the center of the unit cell (indicating that it is not a scanning-related artifact).

As we shall see, the possible structure of the Sb- (2×8) surface is more apparent after examining the (2×4) reconstruction produced by annealing the (2×8) in vacuum. At low magnification the Sb-terminated (2×4) surface looks very similar to the clean, As-terminated GaAs(001)- $\beta 2(2 \times 4)$, as shown in Fig. 3a, with good long-range order and a low defect density. At higher resolution (Fig. 3c, d), though, it becomes apparent that the reconstruction has a different structure than on the clean surface. Even under optimal imaging conditions, each Sb- (2×4) row structure is wider and each 'groove' between rows narrower than observed on the clean $\beta 2(2 \times 4)$. Moreover, on the Sb-containing surface the dimer-like features occur with two distinct heights, 1.5 and 2.5 Å above the lowest point in the unit cell, with approximately two-thirds of the dimers at the larger height. High-pass filtering (Fig. 3d) makes it clear that each (2×4) unit cell includes *three* dimers (with each dimer bond oriented along $[\bar{1}10]$, as usual). On

average, two of these are 'bright' dimers and one is 'dim' in each unit cell, with different permutations randomly occurring on the surface. Given that Sb has a 15% larger covalent radius than As and valence orbitals energetically closer to the Fermi level, the simplest interpretation of these features is that the 'bright' dimers are Sb₂ and the 'dimmer' ones As₂.

Given the 0.5 ML of Sb on the Sb- (2×4) surface, the atomic-resolution STM images lead directly to a simple structural model. As illustrated in Fig. 4a, we propose that the Sb- (2×4) structure is a simple variant of the so-called $\beta(2 \times 4)$ model — three adjacent group-V dimers oriented along $[\bar{1}10]$ on a complete plane of group-III atoms, separated by a missing dimer — once considered for the clean surface [21], but with two-thirds of the surface dimers (0.5 ML) now composed of Sb and the remaining one third (0.25 ML) composed of As. This structure requires the loss of half the As originally on the clean (2×4) surface; a similar displacement of As has been proposed (based on RHEED and XPS) for InAs(001)- (2×4) exposed to Sb [22]. Although almost all dimers appear to be composed of either Sb or As (i.e., both atom-like features within the dimer are a similar height), some height variations on the surface could be due to the occurrence of hybrid Sb–As dimers.

One consequence of having a three dimer (2×4) unit cell is that it requires the replacement of the 0.25 ML of Ga atoms initially missing from the Ga surface layer within the (2×4) reconstruction (see Fig. 1). There are two obvious sources for the required 0.25 ML of atoms: Ga adatoms detached from the surface when the sample is heated to desorb Sb; or in the absence of such Ga mobility, As and/or Sb atoms substitutionally-bonded in Ga sites (As_{Ga}, Sb_{Ga}). Note that if Sb_{Ga} is a significant structural element, either the absolute coverage determination or the obvious interpretation of the STM images must be incorrect. To our knowledge, no previous structural analysis of the Sb-induced GaAs(001)- (2×4) surface has considered a mixture of As and Sb dimer termination. Previous discussion has generally been focused on one, two, or three Sb dimer configurations, with recent results favoring two surface Sb dimers [9,11–

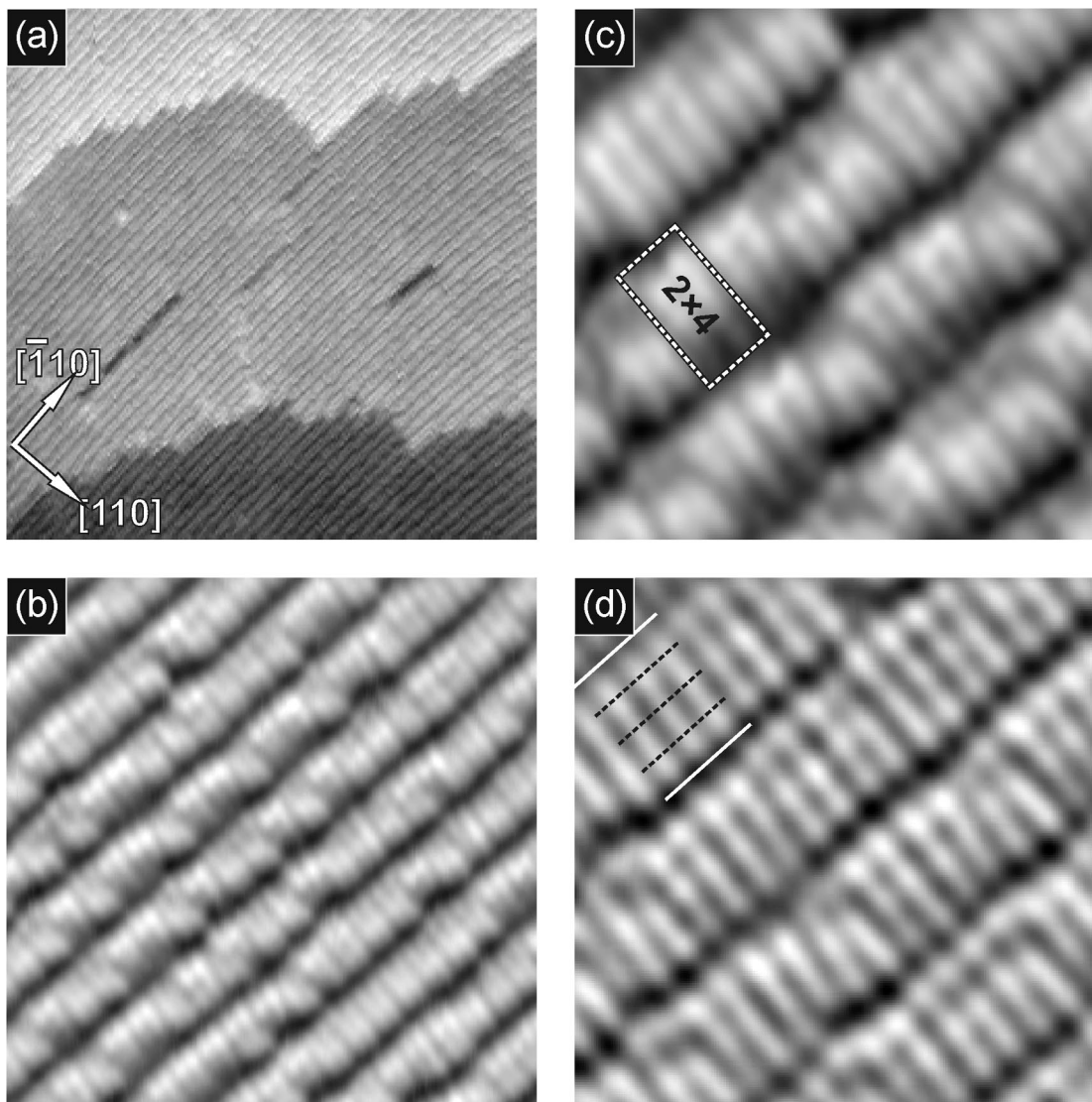


Fig. 3. Filled-state, gray-scale images of the (2×4) -reconstructed surface created by annealing an $\text{Sb}-(2 \times 8)$ surface. The image sizes and tunneling conditions were as follows: (a) 74 nm, 2.5 V, 1.4 nA; (b) 13 nm, 3.0 V, 70 pA; (c) 6 nm, 3.2 V, 0.1 nA. (d) The image shown in (c) after high-passed filtering to accentuate the local height variations. The lines indicate the spacing of the underlying GaAs lattice.

13,15,17]. It will be interesting to see if the mixed, three-dimer structure provides a better fit to the previous experimental data and a lower total energy in first-principles calculations. First-principles calculations could also verify our structural interpretation of the STM images.

Considering the likely structure of the $\text{Sb}-(2 \times 4)$ reconstruction and the Sb coverages, we also tenta-

tively suggest a related model for the (2×8) reconstruction, shown in Fig. 4b, composed of a multilayered mixture of As dimers, Sb dimers, and Sb in substitutional Ga sites. Because each (2×8) unit cell spans two $\beta 2(2 \times 4)$ units, as for the $\text{Sb}-(2 \times 4)$ something must fill in for the missing 0.25 ML of surface Ga. Experimentally, a (2×8) RHEED pattern appears rapidly upon Sb_4 expo-

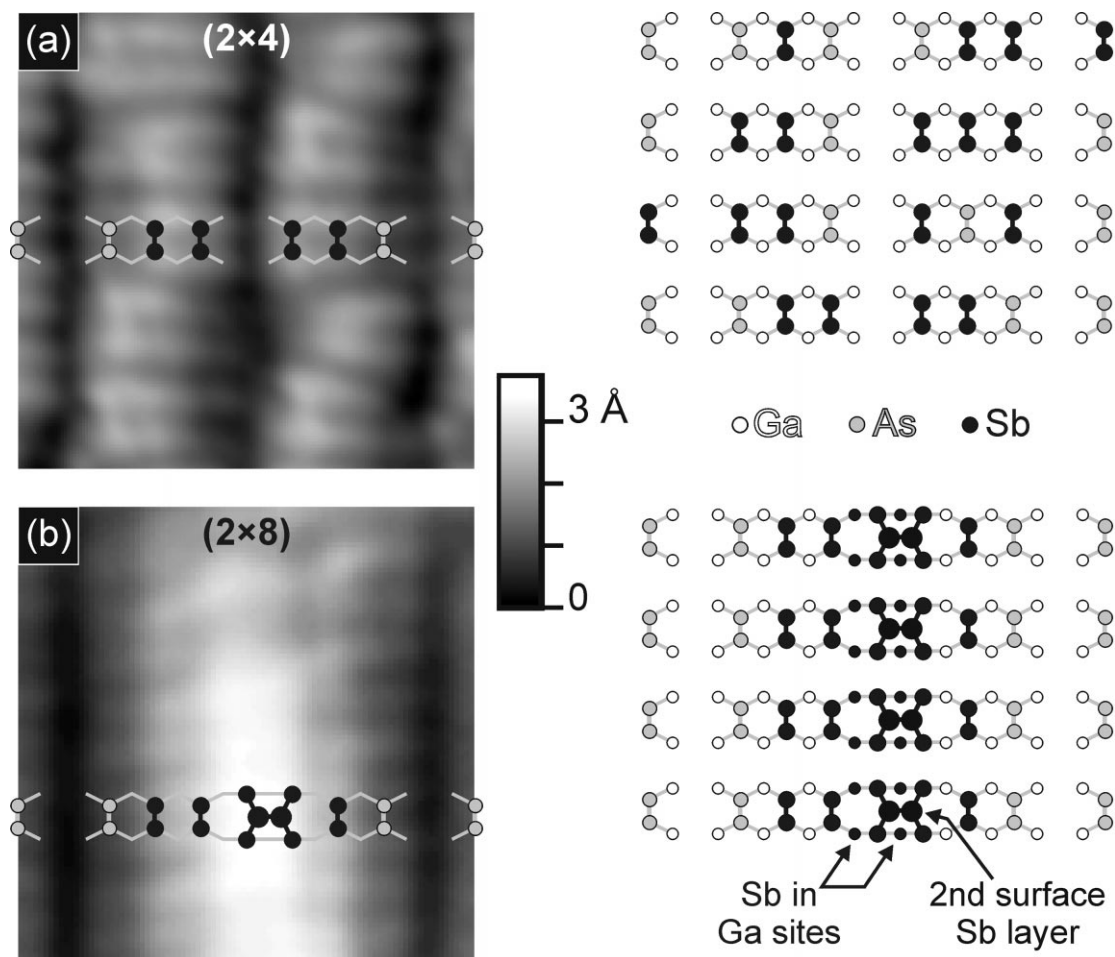


Fig. 4. Proposed models for the Sb-induced (a) (2×4) and (b) (2×8) reconstructions consistent with both the STM images and Sb coverages. Note that the absolute gray scale is set to be the same for both images to facilitate comparison of the topography. The images, $4 \text{ nm} \times 4 \text{ nm}$, were cropped from Figs. 2c and 3c.

sure, suggesting that the transition from the As-terminated $\beta 2(2 \times 4)$ does *not* involve significant mass transport; therefore, in this case the more likely candidate for filling in the missing Ga row is Sb (Sb_{Ga}). On top of this mixed Ga+Sb lattice plane, the three-tiered row structure must be accounted for. Although the heights suggest a three layer structure, the unidirectional dimer-bond orientation within the two lower tiers requires them to be on the same lattice plane (bond angles rotate 90° from plane-to-plane on this zinc-blende surface). The most significant evidence regarding the structure of these two tiers is

that their relative heights and overall appearance are identical to that observed for the 2-Sb-dimers + As-dimer combination on the Sb- (2×4) surface (compare Fig. 4a and b). Based on this similarity, we suggest that the lower dimer tiers and upper dimer tiers are As and Sb, respectively, analogous to the Sb- (2×4) structure. To complete the (2×8) structure, we propose that there is a *second* layer row of Sb dimers on top. Because of the tetrahedral bonding and the odd number of atoms between the grooves, these Sb dimers must, by necessity, be oriented perpendicular to those in the layer below and slightly off-center, as required

by the images. Note that the three-layer Sb structure in the middle of this unit cell is consistent with the propensity of III-Sb surfaces to form reconstructions with multiple surface Sb layers; in fact, this structure is locally identical to a three-layer Sb structure that occurs on GaSb(001) (which has a similar bias-dependent appearance in STM images) [23]. In general, this multilayered structure seems to qualitatively account for the appearance of the images while being consistent with the measured Sb coverage of ~ 1 ML.

Although the proposed Sb-(2×4) model obeys the electron counting rule [21], the model for the (2×8) does not (there are four extra electrons per unit cell). This aspect of the structure is not necessarily a concern, given that weakly metallic GaSb surface reconstructions are stable under similar Sb-rich conditions [23]. It is worth noting, however, that the unit cell could be made charge neutral by replacing two of the substitutional Sb atoms with Ga. But this would reduce the Sb coverage to 0.75 ML, in disagreement with the experimental results. This discrepancy could, in turn, be corrected by replacing some of the As with Sb, but this would further complicate the structure and further reduce its symmetry. A more definitive determination of this structure clearly requires further experimental and theoretical work.

In conclusion, we find that exposing a clean, As-terminated GaAs(001)- $\beta 2(2 \times 4)$ surface to Sb₄ at 490°C produces a (2×8)-reconstructed surface that is terminated with ~ 1 ML of Sb in a complex, but well-ordered structure. We have tentatively proposed a model for this reconstruction incorporating a multilayered mixture of As dimers, Sb dimers, and Sb in substitutional Ga sites. Re-heating an Sb-(2×8) surface to 460°C in vacuum returns the surface to a (2×4) reconstruction with approximately 0.5 ML of the Sb remaining on the surface. Our atomic-resolution STM images clearly show that each Sb-(2×4) unit cell is terminated by *three* dimers, apparently with two-thirds Sb dimers and one-third As dimers in a structure similar to the $\beta 2(2 \times 4)$ model at one time considered for the clean surface. These results contrast with previous proposals that this surface is terminated by one or two Sb dimers.

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References

- [1] T.C. McGill, D.A. Collins, in: R.A. Stradling, J.B. Mullin (Eds.), *Proceedings of the 6th International Conference on Narrow Band Gap Semiconductors*, IOP Publishing, Bristol, 1993, p. S1.
- [2] B.R. Bennett, R. Magno, B.V. Shanabrook, *Appl. Phys. Lett.* 68 (1996) 505.
- [3] P.M. Thibado, B.R. Bennett, M.E. Twigg, B.V. Shanabrook, L.J. Whitman, *J. Vac. Sci. Technol. A* 14 (1996) 885.
- [4] B.R. Bennett, B.V. Shanabrook, P.M. Thibado, L.J. Whitman, R. Magno, *J. Crystal Growth* 175–176 (1997) 888.
- [5] F. Hatami, M. Grundmann, N.N. Ledentsov, F. Heinrichsdorff, R. Heitz, J. Böhrer, D. Bimberg, S.S. Ruvimov, P. Werner, V.M. Ustinov, P.S. Kop'ev, Z.I. Alferov, *Phys. Rev. B* 57 (1998) 4635.
- [6] J.J. Zinck, E.J. Tarsa, B. Brar, J.S. Speck, *J. Appl. Phys.* 82 (1997) 6067.
- [7] R. Kaspi, D.C. Reynolds, K.R. Evans, E.N. Taylor, in: H. Goronkin, U. Mishra (Eds.), *Compound Semiconductors 1994, Proceedings of the 21st International Symposium*, IOP Publishing, Bristol, 1995, p. 57.
- [8] S. Schintke, U. Resch-Esser, N. Esser, A. Krost, W. Richter, B.O. Fimland, *Surf. Sci.* 377–379 (1997) 953.
- [9] F. Maeda, Y. Watanabe, M. Oshima, *Phys. Rev. B* 48 (1993) 14733.
- [10] M. Sugiyama, S. Maeyama, F. Maeda, M. Oshima, *Phys. Rev. B* 52 (1995) 2678.
- [11] P. Moriarty, P.H. Beton, Y.-R. Ma, M. Henini, D.A. Woolf, *Phys. Rev. B* 53 (1996) R16148.
- [12] N. Esser, A.I. Shkrebtii, U. Resch-Esser, C. Springer, W. Richter, W.B. Schmidt, F. Bechstedt, R. Del Sole, *Phys. Rev. Lett.* 77 (1996) 4402.
- [13] T.-L. Lee, M.J. Bedzyk, *Phys. Rev. B* 57 (1998) R15056.
- [14] R. Ludeke, *Phys. Rev. Lett.* 39 (1977) 1042.
- [15] W.G. Schmidt, F. Bechstedt, *Surf. Sci.* 377–379 (1997) 11.
- [16] W.G. Schmidt, F. Bechstedt, *Phys. Rev. B* 55 (1997) 13051.
- [17] G.P. Srivastava, S.J. Jenkins, *Surf. Sci.* 377–379 (1997) 23.
- [18] L.J. Whitman, P.M. Thibado, F. Linker, J. Patrin, *J. Vac. Sci. Technol. B* 14 (1996) 1870.
- [19] E. Kneedler, P.M. Thibado, B.T. Jonker, B.R. Bennett, B.V. Shanabrook, L.J. Whitman, *J. Vac. Sci. Technol. A* 14 (1996) 3193.
- [20] B.R. Bennett, B.V. Shanabrook, M.E. Twigg, *J. Appl. Phys.* 85 (1999) 2157.
- [21] Q.-K. Xue, T. Hashizume, T. Sakurai, *Prog. Surf. Sci.* 56 (1997) 1 and references therein.
- [22] D.A. Collins, M.W. Wang, R.W. Grant, T.C. McGill, *J. Vac. Sci. Technol. B* 12 (1994) 1125.
- [23] L.J. Whitman, P.M. Thibado, S.C. Erwin, B.R. Bennett, B.V. Shanabrook, *Phys. Rev. Lett.* 79 (1997) 693.